

Memory effects in photoinduced femtosecond magnetization rotation in ferromagnetic GaMnAs

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We report a photoinduced femtosecond change in the magnetization direction in the ferromagnetic semiconductor GaMnAs, which allows for the detection of a four-state magnetic memory on the femtosecond time scale. The temporal profile of the magnetization exhibits a discontinuity that reveals two distinct temporal regimes, marked by the transition from a carrier-mediated nonthermal regime within the first 200 fs to a thermal, lattice-heating picosecond regime. © 2009 American Institute of Physics. [DOI: 10.1063/1.3058765]

Magnetic materials displaying *carrier-mediated* ferromagnetic order offer fascinating opportunities for nonthermal, potentially *femtosecond* manipulation of magnetism. Mn-doped III-V ferromagnetic semiconductors are an example.¹ On one hand, their magnetic properties display a strong response to excitation with light or electrical gate and current via carrier density tuning.^{2–4} On the other hand, the strong coupling (~ 1 eV in GaMnAs) between carriers (holes) and Mn ions, inherent in carrier-mediated ferromagnetism, could enable a femtosecond cooperative magnetic response induced by photoexcited carriers. The existence of a femtosecond coherent regime characterized by light-induced magnetization relaxation and collective spin rotation in (III,Mn)V was predicted theoretically.⁵ In addition, a coherent mechanism driving femtosecond spin rotation via *virtual* photoexcitation has recently been demonstrated in antiferro- and ferrimagnets.⁶ Nevertheless, all prior studies of photoexcited magnetization rotation in (III,Mn)V showed dynamics on the picosecond time scale, which accesses the quasithermal lattice-heating regime.⁷ Up to now in these materials, the main observation on the femtosecond time scale has been photoinduced demagnetization.^{8–11}

Custom-designed (III,Mn)V hetero- and nanostructures show rich magnetic memory effects. One prominent example is GaMnAs-based four-state magnetic memory, where “giant” magneto-optical and magnetotransport effects allow for ultrasensitive magnetic memory readout.¹² However, all detection schemes demonstrated so far have been static measurements.

In this letter, we report on photoinduced femtosecond collective magnetization response that allows for femtosecond detection of magnetic memory in GaMnAs. Our time-resolved magneto-optical Kerr effect (MOKE) technique directly reveals a photoinduced four-state magnetic hysteresis via a quasi-instantaneous magnetization rotation. We observe

for the first time a distinct initial temporal regime of magnetization tilt within the first ~ 200 fs, triggered by nonlinear processes without assistance from either lattice heating or demagnetization.

The main sample studied was grown by low-temperature molecular beam epitaxy and consisted of a 73 nm $\text{Ga}_{0.925}\text{Mn}_{0.075}\text{As}$ layer on a 10 nm GaAs buffer layer and a semi-insulating GaAs [100] substrate. The Curie temperature and hole density were 77 K and $3 \times 10^{20} \text{ cm}^{-3}$, respectively. As shown in Fig. 1, our structure exhibits a four-state magnetic memory functionality. Indeed, by sweeping an external magnetic field B , one can sequentially access four magnetic states, $X+ \rightarrow Y- \rightarrow X- \rightarrow Y+$, via abrupt 90° magnetization (\mathbf{M}) switchings between the XZ and YZ planes [Fig. 1(a)]. These multistep magnetic switchings manifest themselves as abrupt jumps in the four-state hysteresis in the Hall magnetoresistivity ρ_{Hall} [Fig. 1(b)] (planar Hall effect).¹² Figures 1(c) and 1(d) show the B scans in the vicinity of 0 T, with the field turning points between the coercivity fields, i.e., $B_{c1} < |B| < B_{c2}$. This leads to a “minor” hysteresis loop, which accesses two magnetic memory states at $B=0$: $X-(0)$ and $Y+(0)$.

We observed transient magnetic phenomena by time-resolved MOKE spectroscopy⁴ using 100 fs laser pulses. The linearly polarized UV pump beam was chosen at 3.1 eV for reasons discussed below. The peak fluence was $\sim 10 \mu\text{J}/\text{cm}^2$. A near infrared beam at 1.55 eV, kept nearly perpendicular to the sample ($\sim 0.65^\circ$ from the normal), was used as probe. The signal measured in this polar geometry reflects the out-of-plane magnetization component M_z .

Figure 2 shows the B field scan traces of the photoinduced change $\Delta\theta_K$ in the Kerr rotation angle at three time delays $\Delta t = -1$ ps, 600 fs, and 3.3 ps. The magnetic origin of this femtosecond MOKE response¹³ was confirmed by control measurements showing a complete overlap of the pump-induced rotation (θ_k) and ellipticity (η_k) changes (left inset in Fig. 2). $\Delta\theta_K$ is negligible at $\Delta t = -1$ ps. However, a mere $\Delta t = 600$ fs after photoexcitation, a clear photoinduced four-

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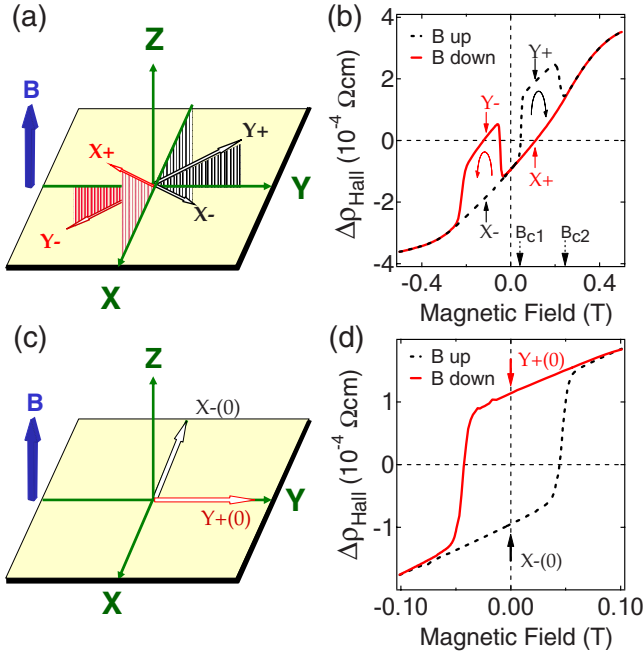


FIG. 1. (Color online) Static magnetic memory. [(a) and (b)] Sweeping a slightly tilted B field (5° from the Z axis and 33° from the X axis) up (dashed line) and down (solid line) leads to consecutive 90° magnetization switchings between the XZ and YZ planes, manifesting as a "major" hysteresis loop in the Hall magnetoresistivity. [(c) and (d)] Minor hysteresis loop with B field sweeping in the vicinity of 0 T. The magnetic memory state $X-(0)$ or $Y+(0)$ is parallel to one of the easy axis directions in the XY plane.

state magnetic hysteresis is observed in the magnetic field dependence of $\Delta\theta_K$ (and therefore ΔM_z), with four abrupt switchings at $|B_{c1}|=0.074$ T and $|B_{c1}|=0.33$ T due to the magnetic memory effects. As marked by the arrows in Fig. 2, the four magnetic states $X+$, $X-$, $Y-$, and $Y+$ for $|B|=0.2$ T

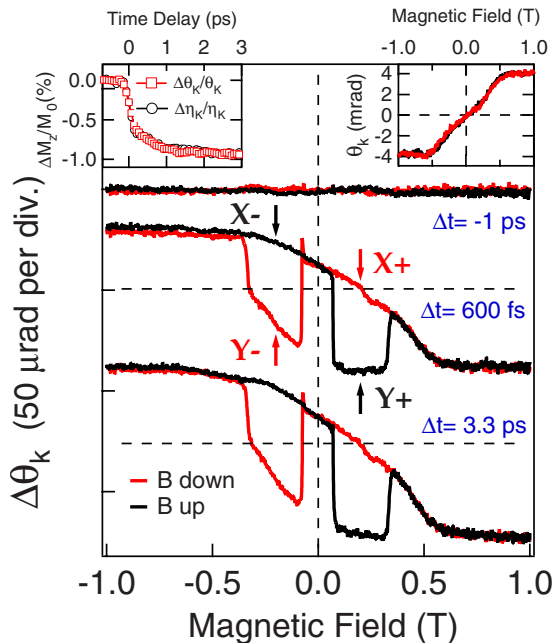


FIG. 2. (Color online) Photoinduced femtosecond four-state magnetic hysteresis. B field scans of $\Delta\theta_K$ at 5 K for time delays $\Delta t = -1$ ps, 600 fs, and 3.3 ps. The traces are vertically offset for clarity. Inset (left): temporal profiles of normalized Kerr (θ_K) and ellipticity (η_K) angle changes at 1.0 T. Inset (right): static magnetization curve at 5 K, measured in the same experimental condition (but without the pump pulse).

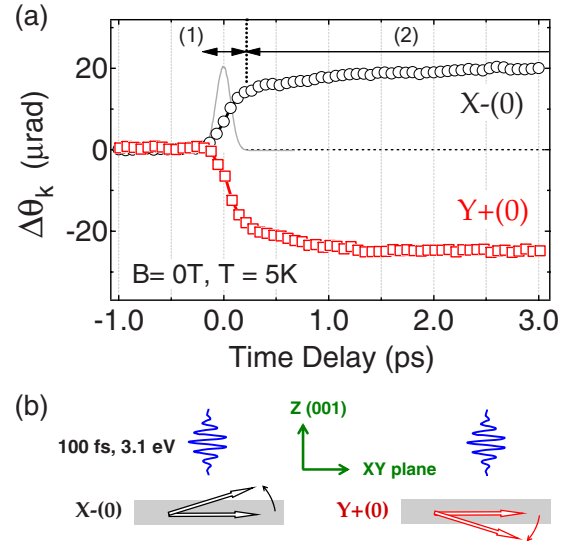


FIG. 3. (Color online) (a) Photoinduced $\Delta\theta_K$ for two in-plane magnetic memory states, shown together with the pump-probe cross correlation (shaded). The opposite, out-of-plane \mathbf{M} rotations for the $X-(0)$ and $Y+(0)$ are illustrated in (b).

give different photoinduced $\Delta\theta_K$. It is critical to note that the steady-state MOKE curve, i.e., θ_K without pump field, does not show any sign of in-plane magnetic switching or memory behavior (right inset in Fig. 2); these exclusively arise from the pump photoexcitation. We note that the photoinduced hysteresis loops at $\Delta t = 3.3$ ps and 600 fs have similar shapes and amplitudes, which confirms that the dynamic magnetic processes responsible for the abrupt switchings occur on a femtosecond time scale.

The photoinduced dynamics of the zero- B field memory states [Fig. 1(c)] elucidates the salient features of the responsible femtosecond magnetic processes. Figure 3(a) shows the temporal profiles of the photoinduced $\Delta\theta_K$ for $X-(0)$ and $Y+(0)$ initial states. Since the initial magnetization vector lies within the sample plane, $\Delta\theta_K$ in the first 200 fs reveals an out-of-plane spin rotation, with negligible contribution from demagnetization (amplitude decrease). More intriguingly, \mathbf{M} in the $X-$ and $Y+$ initial states rotates to different Z -axis directions, as illustrated in Fig. 3(b). This leads to opposite signs of the photoinduced signals and is responsible for the four-state magnetic switchings. Furthermore, the observation of an initial discontinuity in the temporal profiles of the \mathbf{M} rotation reveals *two distinct temporal regimes*, marked in Fig. 3(a): a substantial magnetization rotation concludes after the first 200 fs and is followed by a *much slower* rotation change afterward on the picosecond time scale.

We now discuss the origin of the observed femtosecond spin rotation of the magnetic memory states. In the previously held picture, the photoexcitation alters the anisotropy fields via quasiequilibrium mechanisms, such as heating of the lattice (magnetocrystalline anisotropy) or heating of the spins (shape anisotropy).^{14,15} Since the in-plane magnetic memory states in Fig. 1(c) have negligible shape anisotropy, a significant photoinduced B field within the standard picture can only occur on a time scale of several picoseconds via the lattice heating mechanism and cannot explain the observation of a distinct femtosecond regime. However, it has been shown theoretically^{5,16} that the Mn spin in GaMnAs can respond quasi-instantaneously to the transient hole spin excited

via second-order nonlinear optical processes assisted by strong interactions between Mn and holes and by hole spin relaxation. The resulting magnetization dynamics may be interpreted in terms of light-induced magnetization precession and relaxation.

For photoexcitation close to the Γ point of the valence band (~ 1.5 eV photons), the experimental results only show magnetization dynamics on the picosecond time scale triggered by quasithermal changes in the magnetic anisotropies.⁷ This is in contrast to photoexcitation at ~ 3.1 eV, i.e., close to the L -point where femtosecond magnetic rotation was clearly observed. To understand this qualitative difference and shed light into the microscopic origin of the femtosecond magnetic dynamics, we note that the carrier states differ substantially between the Γ and L points due to the GaAs band structure. A large number of interband transitions with similar energies can be excited for photon energies of ~ 3.1 eV, where the conduction and valence band dispersions are almost parallel. The photoexcited states then cover almost half of the Brillouin zone along the $[111]$ and equivalent directions, i.e., have large momenta while the spin of these band eigenstates is almost parallel to the momentum vector.¹⁶ The Mn-hole spin exchange interaction is also believed to be enhanced along $[111]$ due to strong p - d orbital hybridization.¹⁷ Finally, by mixing the heavy and light hole states, the spin orbit interaction leads to strong hole spin dephasing, which is important for determining the magnetization trajectory during femtosecond time scales.^{5,16} In contrast, ~ 1.5 eV photons excite states with small momenta close to the Γ point, which can be described with the kp effective mass approximation.¹⁸

Chovan and Perakis¹⁶ showed using a simple isotropic two-band model Hamiltonian that the photogenerated effective magnetic field pulse that triggers magnetization rotation and relaxation is determined by the interband optical polarizations. As a result, it depends on the magnetic exchange interaction, optical selection rules, and hole spin dephasing. The valence band structure at the energies of interest therefore plays an important role in determining the trajectory of the magnetization during femtosecond time scales. Our experimental results indicate the photoexcitation of an effective magnetic field ΔB_c along the $[1\bar{1}0]$ direction, which exerts a spin torque $\Delta \vec{B}_c \times \vec{M}$ on \vec{M} that pulls it away from the sample plane. The directions of the above spin torques for the $X-(0)$ and $Y+(0)$ states are opposite, leading to opposite \vec{M} paths (Fig. 3). The quick termination of the initial magnetization tilt implies that ΔB_c decays within the first hundreds of femtoseconds, which suggests that it is limited only by the pulse duration.

In conclusion, we report on the femtosecond magnetic response of GaMnAs, which allows for femtosecond detec-

tion of four-state magnetic memory. Our observations unequivocally identify a *nonthermal*, carrier-mediated mechanism of magnetization dynamics. They clearly reveal a complex scenario of collective magnetization tilt marked by the transition from a nonequilibrium, carrier-mediated regime (< 200 fs) to a picosecond thermal, lattice-heating regime.

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